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INTERACTION OF LIGHT AND SOUND

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INTERACTION OF LIGHT AND SOUND

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ABSTRACT. Gas lasers are used to measure the velocity and absorption of longitudinal hypersonic waves. It is concluded that excitation of hypersound by thermal stresses produced by absorption of short light impulses holds great promise for the study of hyperacoustic properties of a solid body at room temperature.

The area of physics which studies the interaction of light and sound is developing very rapidly at the present time, when laser light sources are used in laboratory studies. Scientific research in this area was initiated by A. Einstein, L. I. Mandel'shtam and P. Debye. In 1910 Einstein [1] calculated the intensity of light scattered by density fluctuations, and Mandel'shtam [2] established the intensity of light scattered due to a fluctuating nonuniformity of the surface of a substance. For the calculations they both analyzed the fluctuations in a three-dimensional Fourier series, and found the intensity from amplitude expansion. /874*

In 1912, completing the Einstein theory of heat capacity of a solid body, Debye [3] advanced the idea that the thermal, kinetic energy of a solid body may be regarded as the energy of normal oscillations (waves) of this body.

From a distance of 60 years, it can be seen that Einstein [1] and Mandel'shtam [2] did not discuss the problems of heat capacity, just as Debye [3] did not deal with problems of light scattering. This now seems almost unbelievable, but at that time this relationship was not understood, and more than six years passed before Mandel'shtam [4] and Brillouin [5] reached the conclusion (independently from each other) that the Einstein and Mandel'shtam Fourier components and Debye waves were the same. From this new point of view, light scattered by adiabatic density fluctuations must be regarded as diffraction of excited light by Debye thermal waves. The scattered light is simulated by elastic waves, which leads to a change in its spectral composition.

If, from the enormous number of Debye waves, we select one, of the frequency Ω with the wave vector **q**, on which a parallel bundle of light falls with the wave vector k_0, and if the scattered light has the wave vector k' and is observed at the angle θ, then in the direction **k'** there will be a maximum of scattered (diffracted) light, if the following Bragg condition is satisfied

$$\mathbf{k}_0 - \mathbf{k}' = \pm \mathbf{q}. \tag{1}$$

As a result of the modulation of the scattered light of the Debye wave frequency Ω, discrete components of the Mandel'shtam-Brillouin (KMB) will be observed

$$\frac{\Omega}{\omega_0} = \frac{\Delta \nu}{\nu} = \pm 2n \frac{v}{c} \sin \frac{\theta}{2}. \tag{2}$$

Measuring $\Delta\nu$ and knowing all the remaining quantities in (2), we may find the speed of hypersound v of the frequency Ω[(1)].

The phenomenon of a thin structure was discovered by Mandel'shtam, Landsberg and Gross in 1930. Two years later Lucas and Biquard [6] and Debye and Sears [7] observed the diffraction of a light wave by an ultraviolet wave. /875
In terms of its nature, this phenomenon was identical to light diffraction by a Debye wave. There was only a difference in the significant amplitude of the ultrasonic wave. The features of light diffraction by ultrasound have been studied in detail (see, for example, [8]). This form of the interaction between light and sound has been included in acoustic and optical studies, and will now apparently play a very important role in the study of the sonic

[(1)] For $n = 1.5$; $\theta = 180°$; $\lambda = 6 \cdot 10^{-5}$ cm and $v = 1.5 \cdot 10^5$ cm$\cdot$sec^{-1}, $\Omega = 2 \cdot 10^{10}$ rad$\cdot$sec^{-1}.

instability in semiconductor-piezoelectrics.

The use of laser light sources has revealed new phenomena in the interaction between light and sound, like the fine structure of the wing of the Rayleigh line (TSK), the force scattering of Mandel'shtam-Brillouin [12, 13] VRMB. Due to powerful laser rays of light, it has been possible to transform the light energy into sonic energy in the ultrasonic frequency range in liquid [14] and crystalline media [15].

Measurements of the Velocity and Absorption of Longitudinal Hypersonic Waves

The use of gas lasers has made it possible to use the position of the KMB [Formula (2)] to measure the speed of hypersound within an accuracy of fractions of a percent, and the use the width of the components to find the hypersound absorption coefficient within an accuracy of several percents. After the first such study with a laser source [16], there was an entire group of studies, which is still constantly increasing. Certain previous measurements on the speed of sound dispersion were refined, and data were obtained for other substances.

The simultaneous determination of the velocity and absorption of hypersound made it possible to directly check experimentally the formula of the relaxation theory. It was found that it completely corresponds to experience for several cases. For the example shown in Figure 1, the corresponding curve was found, which was obtained from low frequency and hypersonic measurements, /876
and dots were placed in the interval, obtained primarily by Berdyyev, Lezhev and others [17]. The formulas with one τ do not in every case closely describe the behavior of this dependence. In the case of viscous liquids, the relaxation theory is generally inapplicable [18].

The propagation of sound in viscous media is described by a non-local theory, developed by Isakovich and Chaban [19]. A whole group of such problems

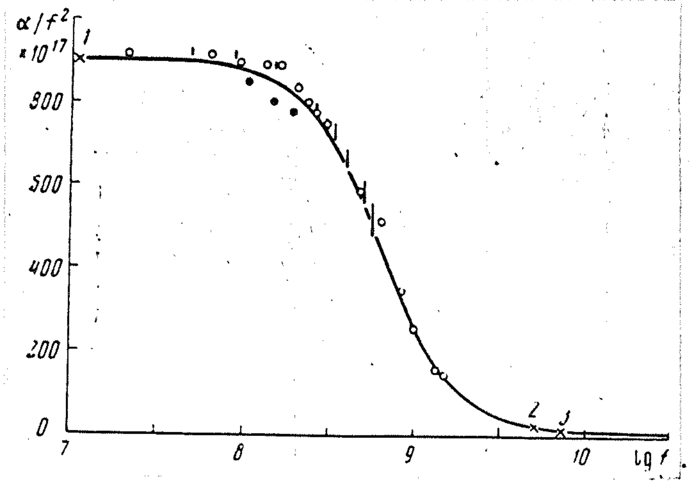

Figure 1. Dependence of α/f^2 on the frequency f in benzene. The solid curve is based on the formula of the relaxation theory, based on the low frequency point (1) and the high frequency point (2) found from the fine structure of the Rayleigh line during excitation with a laser. Point (3) was obtained during the excitation of the mercury spectrum line. The other points are experimental points obtained by different authors from ultrasound measurements.

is of great interest, but we cannot discuss them in detail, and, by way of an example, shall only give one of the many curves for the dependence of the speed of sound on temperature in triacetin (Figure 2). This field of research continues to develop successfully, and has now been extended to the range of frequencies from 10^{-2} to $2 \cdot 10^9$ Hz, and from viscosities from fractions of a power to the viscosity of a vitreous state.

Propagation of Transverse Hypersonic Waves

In addition to polarized light of a Rayleigh triplet, in the spectrum of scattered light there is light which is depolarized, caused by anisotropy fluctuations — the wing of the Rayleigh line. In certain liquids, such a narrow, central part of the wing can be seen that it can be separated from the remaining wide part of the wing only by using interference spectroscopy [12].

The new phenomenon which we have discovered [9] consists of the fact that this narrow part of the wing in x-polarization (I_{zx}) represents a doublet, and is not a continuous distribution of intensity, as was previously assumed. We have explained this phenomen by the fact that light, scattered due to anistropy fluctuations, is modulated by the Fourier component of the deformation fluctuations, corresponding to the Bragg condition or, in other words,

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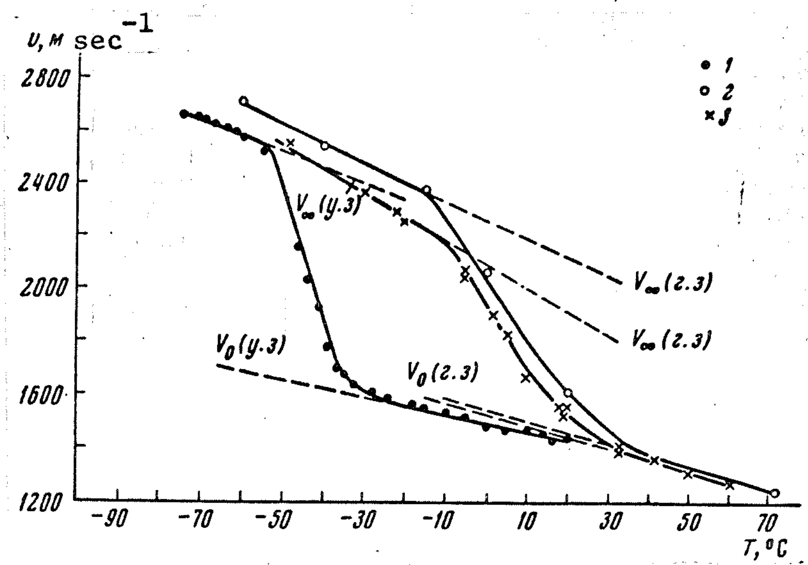

Figure 2. Dependence of ultrasound and hypersound velocity on the temperature in triacetin. The solid curve is theoretical [19]; the experimental results are plotted by points: 1 - ultrasound; 2 - hypersound (thermal scattering of light); 3 - hypersound (forced scattering of light).

it is modulated by a transverse hypersonic wave. If this is tuue, then the phenomenon may be described by formulas of the well-known theory of Leontovich /877 [12, 20].

$$I_{zx}(\omega)=I_{yz}(\omega)=\mathrm{const}\left\{\frac{\omega^2\tau}{\omega^2+\tau^2(\omega^2-\Omega_T^2)^2}+\frac{\tau}{1+\omega^2\tau^2}\right\}. \quad (3)$$

$$I_{yx}(\omega)=\mathrm{const}\left\{\frac{(\omega^2-\Omega_L^2)^2\tau}{(\omega^2-\Omega_L^2)^2+\omega^2\tau^2(\omega^2-\Omega_S^2)^2}+\frac{\tau}{1+\omega^2\tau^2}\right\}, \quad (4)$$

$$\Omega_S=2n\omega_0\frac{v_S}{c}\sin\frac{\theta}{2};\qquad \Omega_T=2n\omega_0\frac{v_T}{c}\sin\frac{\theta}{2} \quad (5)$$

$$\Omega_L^2=\Omega_S^2-\frac{4}{3}\Omega_T^2;\qquad v_S=\left(\frac{\lambda+\frac{4}{3}\mu}{\rho}\right)^{1/2};$$
$$v_T=(\mu/\rho)^{1/2}. \quad (6)$$

Here ω is the frequency computed from the frequency of the excited light ω_0; v_S and v_T — phase velocities of transverse and longitudinal waves; θ — scattering angle; μ — displacement modulus. The remaining notation is the customary notation [12].

The Leontovich theory [20] assumes, for purposes of simplicity, that the relaxation of a shear viscosity τ is the same as the relaxation time of anisotropy, although generally speaking, these times are different — as was apparent from the very beginning to the author of the theory. It would then (almost 30 years later) be important to find the general rules, and simplify the theory. Now, when we have discovered very exact phenomena in the scattering spectrum, it is necessary to call attention to the difference in the times.

Analyzing (3), we can see that the first term in the formula indicates two maxima at a frequency of $\omega \pm \Omega_T$, and the second term — a maximum at the frequency $\omega = 0$. All three maxima have the same intensity. In several liquids, such as nitrobenzene, quinoline, anoline, salol, benzophenon, and others, in the narrow part of the wing a more or less clear picture of the doublet may be observed, with a rather small gap between them. This phenomenon was observed in the experiments of Stigman and Stoicheff [21] and in other unpublished studies. In our experiments [9, 22, 23] and the experiments of [21] it was found that, in agreement with the requirements of Formula (5), the linear dependence between Ω_r and $\frac{\theta}{2}$ is satisfied. In addition, in our experiments [9, 22, 24] it was shown that a polarization relationship holds, which was predicted by theory (3), i.e., $I_{zx}(\omega) = I_{yz}(\omega)$.

If the Leontovich theory actually describes the observed phenomenon, it may be expected that, with an increase in the liquid viscosity (temperature reduction), the displacement modulus will increase, and consequently the distance between the doublet components must increase. For a certain viscosity, a clear triplet appears. Since damping of transverse hypersound must decrease with an increase in viscosity, KMB must become narrower, and due to an increase in μ the distance between them will increase. Thus, the same polarization relationships are retained as for slightly viscous liquids, where TSK was observed.

Our experiments with glass and melted quartz [24] actually showed that the polarization relationships correspond to the requirement of theory, and the lines of the thin structure remain narrow. Thus, it would appear that everything has been explained by the existing theory. However, this is not the case. The fact is that, when the viscosity increases by an order of magnitude in quinoline, the TSK components either do not change their position, or the distance between them is even reduced [21, 25]. This contradicts the facts mentioned above, and makes it necessary to study the kinetics in the /878 entire interval in which the viscosity changes up to a vitreous state. This research was recently carried out in our laboratory [25]. The research results are given in Figure 3.

The results of the experiment show that in actuality the phenomenon is much more complex than has been assumed. All of our discussions and formulas of the Leontovich theory pertained to the left part of the graph, where the events developed just as we assumed. In the right part of the graph it may be seen that the distance between the doublet components increases when the temperature increases. This behavior of the curve is not given in the Leontovich theory [20]. It was explained that in general the TSK cannot be predicted from any one of the existing theories. This phenomenon will require its own explanation by a new theory. We were able to provide a qualitative explanation by introducing new anistropy relaxation times [25]. To explain these TSK, Volterra [26], Solov'yev and Romanov [27] expanded the Leontovich theory to the case of two anistropy relaxation times and provided a qualitative explanation for the TSK (right branch in Figure 3). In the Rytov theory [28] a scheme with two anistropy relaxation times is composed, and formulas are obtained which describe qualitatively the entire phenomenon (both branches in Figure 3). To provide a quantitative description of the experiment results, along with the relaxation theory a non-local theory should also be used for

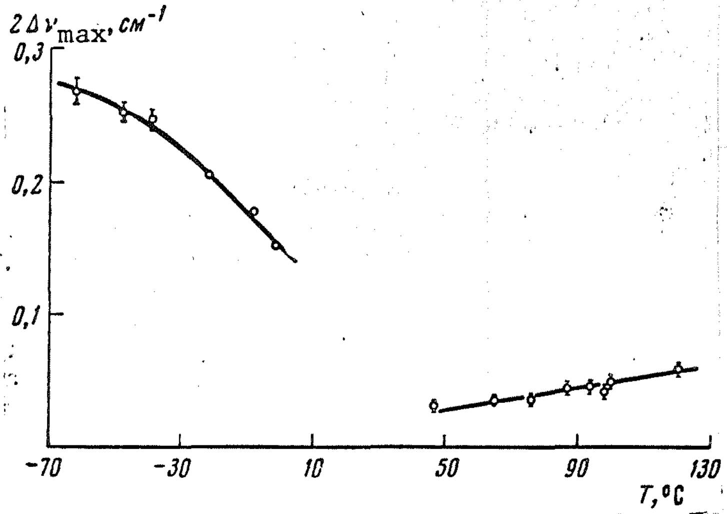

Figure 3. Dependence of the distance between the components of a "transverse" doublet $2\Delta\nu_{max}$ on the temperature in salol.

the case when the medium viscosity is large.

Forced Scattering of Mandel'shtam-Brillouin

The particularly strong interaction between light and sound occurs in the forced scattering process of Mandel'shtam Brillouin (VRMB). This phenomenon consists of the fact that the interaction between the exciting light wave and the Debye thermal wave by means of electrostriction intensifies the Debye wave so greatly that its intensity may amount to megawatts.

If an electric field whose resulting intensity is E acts on the isotropic body, then the striction pressure caused by this field [12] /879

$$P = \left(\rho \frac{\partial \varepsilon}{\partial \rho}\right) \frac{E^2}{8\pi}, \tag{7}$$

$$\begin{aligned} \mathbf{E} = \mathbf{E}_0 \cos[\omega_0 t - (\mathbf{kr})] + \mathbf{E}_1 \cos[(\omega_0 - \Omega)t - (\mathbf{k}_1\mathbf{r})] + \\ + \mathbf{E}_2 \cos[(\omega_0 + \Omega)t - (\mathbf{k}_2\mathbf{r})], \end{aligned} \tag{8}$$

It may be seen from this that the nonzero pressure or the sound wave will be

$$P = \left(\rho \frac{\partial \varepsilon}{\partial \rho}\right) \frac{1}{8\pi} \{E_0 E_1 \cos[\Omega t - (\mathbf{qr})] + E_1 E_2 \cos[\Omega t - (\mathbf{k}_1 - \mathbf{k}_2)\mathbf{r}]\}, \tag{9}$$

This means that the Debye wave, which produced the Mandel'shtam-Brillouin component during scattering at the given angle θ, when the Bragg condition

$k_0 - k_1 = \pm q$ is satisfied, will be intensified by a factor of E_0. This is the mechanism by which sound is intensified due to the energy of a light wave in the process of nonlinear interaction. A more complete examination of the problem requires the concurrent solution of a system of nonlinear Maxwell equations and hydrodynamic equations, if a liquid is being discussed, and equations of elasticity theory, if a solid body is being discussed [12, 29].

It may be seen from an analysis of the solutions that in the stationary case of VRMB, i.e., when the duration of the exciting impulse t is much greater than the lifetime of a phonon $\tau_p (t \gg \tau_p)$ and $I_{MB} \ll I_0$,

$$I_s = I_{MB} \frac{\Omega}{\omega_0} \frac{g_{MB}}{2\alpha}$$

(under the condition $g_{MB} \ll 2\alpha$), where α is the amplitude absorption coefficient of sound and g_{MB} — VRMB amplification factor. In the nonstationary regime, we have

$$I_s = I_{MB} \frac{\Omega}{\omega_0} \frac{vt}{L}$$

(under the conditions $vt \ll L$, L — length of the nonlinear interaction). In the ideal case, when there are no losses and each photon produces one phonon, the maximum intensity of sound is determined by the Menli-Roy relationship

$$I_s = \frac{\Omega}{\omega_0} I_{MB}.$$

It is practically impossible to approach this limit, because at large intensities the sound becomes nonlinear, and losses increase as a result of the transformation of sound with basic frequency into harmonics. The nonlinear nature of hypersound in a quartz crystal at a frequency of $2.6 \cdot 10^{10}$ Hz is apparent at intensities of $3 \cdot 10^2 W \cdot cm^{-2}$ [29]. In experiments with nonocrystalline quartz in our laboratory, the strength of hypersound reached $3 \cdot 10^3$ W, and the intensity reached $MW \cdot cm^{-2}$ [30]. The occurrence of the second hypersound harmonic for VRMB in water was observed experimentally [31]. In our experiments with crystalline quartz at a temperature of 80° K intense longitudinal and transverse hypersonic waves were observed by the appearance of the VRMB.

We must point out that the VRMB represents a generator of intense hypersonic and ultrasonic waves, whose nonlinear properties must be studied by optical and acoustic methods. This region has still not been completely studied.

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Generation of a Sonic Wave during the Nonlinear Interaction of Two Light Waves having Differnet Frequencies

In forced scattering, the nonlinear interaction of two light waves of differing frequency and a Debye wave leads to amplification of the thermal wave existing in the medium. The experiments of Korpel [14] and Coddas [15] established the displacement of two light waves, whose frequency differs by the quantity Ω, and found a sonic wave of the frequency Ω in the medium. In these studies they had to diffract the intense light wave, which was radiated by a laser with modulated quality, by ultrasond in water [14] and in a crystal [15]. As a result, the light which passed directly (zero maximum) had the frequency of the initial light ω_0, and the light which was diffracted at the Bragg angle, had a frequency of $\omega_0 - \Omega$, where Ω is the frequency of the sonic wave.

When both rays or one of them, had great intensity and, consequently, a strong electric field, then just as in the case of VRMB electrostriction arose. According to Formula (9) (where we set $E_2 = 0$) we obtained the striction pressure which is equal to

$$P = \frac{E_0 E_1}{8\pi}\left(\rho \frac{\partial \varepsilon}{\partial \rho}\right)_s \cos[\Omega t - (\mathbf{qr})].$$

Here Ω is the light frequency difference ω_0 and ω_1, and $\mathbf{q}$ equals the difference of the wave vectors $\mathbf{k}_0$ and $\mathbf{k}_1$.

If the light rays are encountered at an angle equal to twice the Bragg angle in the medium, and the phase velocity of the striction wave with a pressure $v_{fr} = \Omega/q$ equals the phase velocity of sound in this medium v, then as a result of the interaction of these light waves a sonic wave develops. The front of this wave will be the bisector of the angle at which the light waves intersect.

Sonic waves of the frequency 45 and 57 mHz were detected in water [14] and with a frequency of 720 mHz in crystals of $SrTiO_3$ and quartz [15]. The intensity of the sound thus produced is determined by the following relationship [15]

$$I_S = FL^2 I_0 I_1, \tag{10}$$

where L is the interaction length; I_0, I_1 — intensity of the interacting bundles of light; F — coefficient equal to the following [15]

$$F = \frac{h^6 p_{ij}^2 C_{ij} \Omega^2}{8c^2 \rho v^3}.$$

Here p_{ij}, C_{ij} are the elastic-optical and elastic constants, respectively.

Due to the small Bragg angle, the interaction region practically equals the width of the light bundle L' at high frequencies. When using short light impulses, the sound cannot intersect the entire width of the bundle of sound, and the region of interaction will equal vt', where t' is the duration of the light impulse. If vt' < L', then Expression (10) must be decreased by $(vt'/L')^2$. In these crystalline samples, the intensity of a sonic wave was ∿ 50 $dB \cdot mm^{-2}$.

It is possible to change the frequency of the light waves not only by light diffraction by ultrasound, but also by any other method — for example, using the VRMB, the forced entropy (temperature) scattering of light, the modulation of light by Kerr and Pokels elements, or simply producing double mode generation of a laser, etc. Depending on the frequency difference of light waves in solid bodies, sufficiently intense sound waves may be produced in a gigacycle frequency range.

Transformation of Light Impulses into Sonic Impulses /881

Almost immediately after solid body lasers were created, it was found that absorption in a liquid and solid body of powerful light impulses leads to the formation of a sonic impulse. White [32] made the most detailed theoretical study of the problem of excitation and transformation of light impulses into sonic impulses. He also studied the problem of the periodic interaction of light impulses and the excitation of a sonic wave. He showed that the transformation of laser impulses into sound holds particularly great promise.

In the periodic interaction of light on an absorbing surface, the amplitude of the sonic wave will be proportional to the square of the amplitude of the light wave, and inversely proportional to the sound frequency [32]. This excitation of sound is extremely effective, because its amplitude is proportional to the peak intensity of a light impulse.

During the absorption of a light impulse in a thin layer of a solid body (metals and semi-conductors, which are not transparent in the region of the emitted light, or are dielectrics on whose surface a metallic layer is applied) enormous temperature gradients arise, and the temperature changes very rapidly. Ready [33] calculated that the nonstationary absorption of an impulse from a ruby laser with an intensity of 17 $MW \cdot cm^{-2}$ and a duration of 25 ns produces a temperature gradient of 10^6 $degree \cdot cm^{-1}$ and a temperature change at the rate of 10^{10} $degree \cdot sec^{-1}$. This absorption effect produces a blast wave. If the light impulses follow each other with a specific frequency, then a periodic blast wave and its harmonics will be observed.

Brienza and DeMaria [34] excited ultraviolet sound impulses and waves using a laser on a neodymium glass with modulated quality and with synchronized modes. In this experiment, hypersound with a frequency of $2 \cdot 10^9$ Hz was excited in lithium niobate, quartz, and sapphire, and recorded at room temperature. This great achievement will no doubt be developed and extended to the excitation of a higher frequency hypersound.

It can be seen that the excitation of hypersound by thermal stresses produced by the absorption of short light impulses holds great promise for the study of hyperacoustic properties of a solid body at room temperature, and also at higher and lower temperatures. This method of the optical excitation of sound will have acoustical applications.

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